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THE DECAY OF ORIENTED Sm^{145} *

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ABSTRACT

Samarium-145 nuclei were oriented at low temperatures in neodymium ethylsulfate in cerium magnesium nitrate lattices. From the temperature-dependent angular distribution of the 61-keV γ ray in Pm^{145}, a magnetic moment $\mu = 0.92 \pm 0.06$ nm was deduced for the ground state of Sm^{145}. The ratio of attenuation coefficients in the 61-keV state of Pm^{145} in the two lattices was found to be $G_2(CMN)/G_2(NES) = +0.44(10)$. Evidence is presented which indicates that temperature gradients in CMN can lead to erroneously low values for magnetic moments.

INTRODUCTION

Radioactive isotopes of most of the rare-earth elements have been oriented in lattices of neodymium ethylsulfate, $Nd(C_2H_5SO_4)_3 \cdot 9H_2O$, (NES), or of cerium magnesium nitrate, $Ce_2Mg_3(NO_3)_{12} \cdot 24H_2O$ (CMN).[1] In a systematic survey of oriented rare-earth nuclei in these lattices started in 1960 we obtained results for Sm^{145} that we could not explain. The magnetic moments derived from studies in the CMN and NES lattices were apparently quite different. While some of the earliest work in the field of nuclear orientation showed discrepancies of this kind, most of these could be attributed to poor experimental technique and would disappear when the work was done more carefully. Our Sm^{145} results were checked for all the known sources of error, however, and refinement of technique only confirmed them. With the recent discovery that the temperature scales in use for both NES and CMN were seriously in error, the Sm^{145} data were corrected to the new scales,[2,3] and the magnetic moments derived from the two salts are now in excellent agreement. The determination of this magnetic moment is described here.

Having fitted the nuclear orientation data to derive moments, one also obtains, for each lattice, A_2, the coefficient of $P_2(\cos \theta)$ in the γ-ray angular distribution function. This A_2 may have the full value $B_2U_2F_2$ implied by angular momentum theory alone, or it may be attenuated by a factor G_2 which describes reorientation in an intermediate state. For this case (i.e., the 61-keV, $t_{1/2}$ = 2.6 nsec state[4] of Pm^{145}), we have found substantial attenuation in the CMN lattice, as described below.

EXPERIMENTAL

The Sm^{145} activity was prepared by neutron irradiation of enriched Sm^{144},

followed by ion-exchange separation from other rare-earth contaminants, notably

(daughter) Pm^{145}, immediately before use. The Pm^{145}/Sm^{145} γ-ray intensity

ratio was easily kept below 1%. As the Pm^{145} γ rays were shown in separate

experiments to be essentially isotropic, no correction was necessary.

The heavier rare earths grow substitutionally into the CMN lattice poorly,

the difficulty increasing with Z. Although samarium is known to go into the double

nitrate lattice,[5] the complete analysis of a nuclear orientation experiment

requires that nearly all of the activity " in the crystal" should in fact be

substitutionally in the trivalent lattice sites. Most of the CMN crystals

that were used in this work were quite clear, though none were completely

clear.There was no tendency for γ-ray anisotropy to increase with crystal

clarity (as would be the case if significant fractions of the activity were

present as inclusions). A discrimination factor of only 10:1 against Sm

going into CMN was determined in a separate experiment. Thus, it seems

unlikely that the low value of A_2 reported below can be attributed to Sm^{145}

that is in the sample but not oriented in lattice sites.

The apparatus has been briefly described elsewhere.[6] It seems worth-

while, however, to discuss here in some detail the factors relating to a very

important parameter in nuclear orientation experiments, the heat leak.

In most cryogenic experiments involving adiabatic demagnetization it

is possible, by isolating the specimen and employing a paramagnetic salt with

a large heat capacity, to reduce the heat leak into the specimen to negligible

proportions. Heat leaks as low as 1 erg/min have been reported although

100 ergs/min is a more common figure.

For single-crystal nuclear orientation work many of the devices used
to reduce the heat leak in other experiments are automatically excluded, and
except in special cases one is forced to think in terms of a total "warm-up time"
(i.e., the time required after demagnetization, for the specimen to reach
the temperature of the helium bath). If one wishes to determine radiation
distributions with any reasonable accuracy, it is essential that the "warm-up
time" be much longer than the counting interval. Inasmuch as meaningful
angular distributions may be obtained only when the entire specimen is at a
nearly uniform temperature (i.e., soon after demagnetization), this implies
that warm-up times of many minutes, or preferably several hours, are highly
desirable.

In order to produce warm-up times of 1-2 hours in a CMN crystal, or ~8
hours in an NES crystal, in a simple apparatus with which it is possible to
go from room temperature to demagnetization temperatures in less than 2 hours,
we have developed the design shown in Fig. 1. At the top (not shown) is a
brass cap, silver-soldered to a tube (with light traps to prevent direct
radiation from above) which acts as a vacuum line and support. A short
tungsten rod is silver-soldered to the inside of the cap, and a 2 mm glass
rod is sealed to its lower end. This glass rod is about 30 cm long and supports
a compressed salt pill of $Mn(NH_4)_2(SO_4)_2 \cdot 6H_2O$, which has a high magnetic heat
capacity and cools, in the stray field of the electromagnet, to about 0.17°K,
where its high surface area adsorbs gas leaking down into the chamber. Below
this is a slurry of $Cr_2K_2(SO_4)_2 \cdot 12H_2O$ and glycerine, which cools to ~0.01°K and
acts as a sink for heat conducted down the glass support rod as well as a
second adsorbent for gas. Finally the CMN crystal is attached to a glass

rod framework with Duco cement. The experimental chamber consists of a glass tube, sealed at the bottom, joined to a short copper tube at the top with a Housekeeper seal.

Assembly of the apparatus consists of attaching the crystal-salt assembly, soldering the chamber on at the top, painting the chamber with Aquadag (colloidal graphite), mounting the inductance coils, and wrapping all in black paper as a precaution against radiation leaks through pinholes in the Aquadag. With this apparatus warm-up times are typically in the 80-100 min range for a 5-10 gram crystal of CMN and ~10 hours for NES.

Temperatures were determined by measuring the susceptibility of the (NES or CMN) crystal and correlating this with the magnetic field and temperature from which demagnetization was done, using the known T-S relations.[2,3] Data for NES and CMN are shown in Figs. 2 and 3. The 61-keV γ ray was detected at $0°$ and $90°$ from the trigonal axis by $3'' \times 3''$ NaI(Tl) detectors. Care was taken to insure that data were recorded only when the crystals were at uniform temperatures, i.e., immediately after demagnetization. Thus each experimental point represents a complete demagnetization run. In Fig. 4 data taken in this way are contrasted to those taken as a CMN crystal warms up from the lowest temperature, the latter being erroneously low.

In the CMN runs the saturation values of $W(\theta=0)$ and $W(\theta=\pi/2)$ were never quite consistent with a $P_2(\cos \theta)$ distribution, the function $W(0) - 1$ being slightly larger than the expected $2(1-W(\pi/2))$. This effect, which was scarcely outside of experimental error, was originally attributed to scattering by the CMN crystals, which were typically 3 mm $\times$ 20 mm $\times$ 20 mm, the short dimension being along the c axis. Extensive tests did not confirm a scattering mechanism, however, and the small deviation remains puzzling.

DISCUSSION

The spin Hamiltonain relevant to this work is

$$\mathcal{H} = AS_z I_z + B(S_x I_x + S_y I_y) \quad .$$

In the double nitrate lattice we have $A > B$. For Sm^{147} in samarium magnesium nitrate,[5] $A = 0.0346(5)$cm^{-1} and $B \leq 0.010$ cm^{-1}. In the ethylsulfate lattice $B > A$: for Sm^{147} in lanthanum ethylsulfate,[7] $A = 0.0060(1)$cm^{-1} and $B = 0.0251(1)$cm^{-1}. For the lowest electronic state of Sm^{3+} in both lattices $S = 1/2$. The nuclear spin of Sm^{147} is $7/2$[5,7]: for Sm^{145} spin $7/2$ is the most probable assignment.[4] Thus rather direct comparisons of A and B for the two isotopes is possible. The ratio A/B is of course the same for both isotopes in a given lattice. In calculating the temperature dependence of the orientation parameters[8] B_k, we therefore have only one independent variable to determine for each lattice.

In this work the angular distributions could be described to first approximation[9] by functions of the form

$$W(\theta) = 1 + A_2 P_2(\cos \theta),$$

where A_2 can be written[10] $A_2 = B_2 U_2 G_2 g_2 F_2$. The solid angle factor g_2 is easily calculated, and B_2 may be determined from the temperature dependence of A_2 and divided out. Reorientation during electron-capture decay is described by U_2. This reorientation may be calculated in principle, but as the details of the decay are not known, we can only set the limits $0.466 \leq U_2 \leq 1.000$, based on the values 1.000, 0.810, and 0.466 for 0, 1, and 2 units of angular

momentum in the lepton system. A value of $U_2 = 0.9 \pm 0.1$ is very likely. The

value of F_2 is also <u>a priori</u> unknown, because it depends on the E2/M1 mixing

ratio of the 61-keV γ ray. In practice, then, we can determine $G_2 U_2 F_2$ for

each lattice, and derive the ratio of attenuation factors G_2: the results of

this analysis are set out in Table I.

The excellent agreement of the values of the magnetic moment derived

from the two lattices supports the usefulness of the CMN lattice for magnetic

moment determinations. In the past the magnetic moments derived from nuclear

orientation measurements in this lattice have almost invariably been erroneously

small. This has been due, in our opinion, to the lack of a reliable tempera-

ture scale, to attenuation in intermediate states, and to high heat leaks. By

minimizing the heat leak it should now be possible, at least in cases for

which the orientation can be saturated, to determine nuclear moments reliably.

The Sm^{145} nuclear moments might be expected to follow either of two

empirical trends, that of 83-neutron nuclei or that of samarium isotopes.

The actual trend is shown in Fig. 5. It is possible that the A-dependence

of μ is smoother than this figure suggests, and perhaps even monotonic, as

$\mu(Ce^{141})$ is based on an extrapolated value of $\langle r^{-3} \rangle$ for 4f electrons.[11] As

this extrapolation is questionable, the value $\mu = 0.97$ nm for Ce^{141} must be

regarded as tentative.

From the derived values of $G_2 U_2 F_2$, we have $G_2(\text{CMN})/G_2(\text{NES}) = +0.44(10)$.

It is tempting to interpret this ratio as suggesting $G_2(\text{NES}) \sim 1$, $G_2(\text{CMN}) \sim 0.4$

This would be consistent with $G_2(\text{CMN})$ being a "hard core" value for time-

independent perturbations in the 61-keV state and with little or no attenua-

tion in NES. It is clear that much <u>more</u> attenuation occurs for this state

in CMN than in NES, and this is consistent with evidence for other cases.[12,13]

Perhaps this difference suggests that changes in oxidation state following

beta decay is faster in CMN, where the rare-earth ion is surrounded by nitrate

ions, than in NES, where water molecules constitute the immediate environment.

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FOOTNOTES AND REFERENCES

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†National Science Foundation Senior Postdoctoral Fellow 1966-7.

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9. In the CMN case only, we found consistently $W(0) - 1 > 2[1-W(\pi/2)]$, suggesting an $A_4 P_4 (\cos \theta)$ term in the angular distribution, with $A_4 \sim 1\%$ at the lowest temperatures. As this would require a larger E2 admixture in the 61-keV γ ray than the K/L ratio[4] allows, we tentatively attribute the effect to scattering in the cryostat.

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Table I. Summary of results.

Lattice	hfs constant	μ_{145}/μ_{147} [a]	$G_2U_2F_2$
NES	$B = 0.028(3)\,\text{cm}^{-1}$	1.12(11)	+0.127(20)
CMN	$A = 0.040(4)\,\text{cm}^{-1}$	1.15(12)	+0.055(8)

[a] Using $\mu_{147} = 0.812$ nm, we have $\mu_{145} = 0.92(6)$ nm.

FIGURE CAPTIONS

Fig. 1. The cryostat.

Fig. 2. Plot of $W(0) - W(\pi/2)$ for the 61-keV γ ray of Pm^{145} following the decay of Sm^{145} oriented in a neodymium ethylsulfate crystal. Theoretical curve is shown.

Fig. 3. Similar to Fig. 2, but for the cerium magnesium nitrate lattice.

Fig. 4. Intensity along the c axis of the 61-keV γ ray following the decay of Sm^{145} oriented in CMN. Open circles were determined with the crystal at a uniform temperature. Closed circles represent data obtained as the crystal warmed up: these data give a magnetic moment that is 20% low.

Fig. 5. Magnetic moments of Sm^{145} and related nuclei.
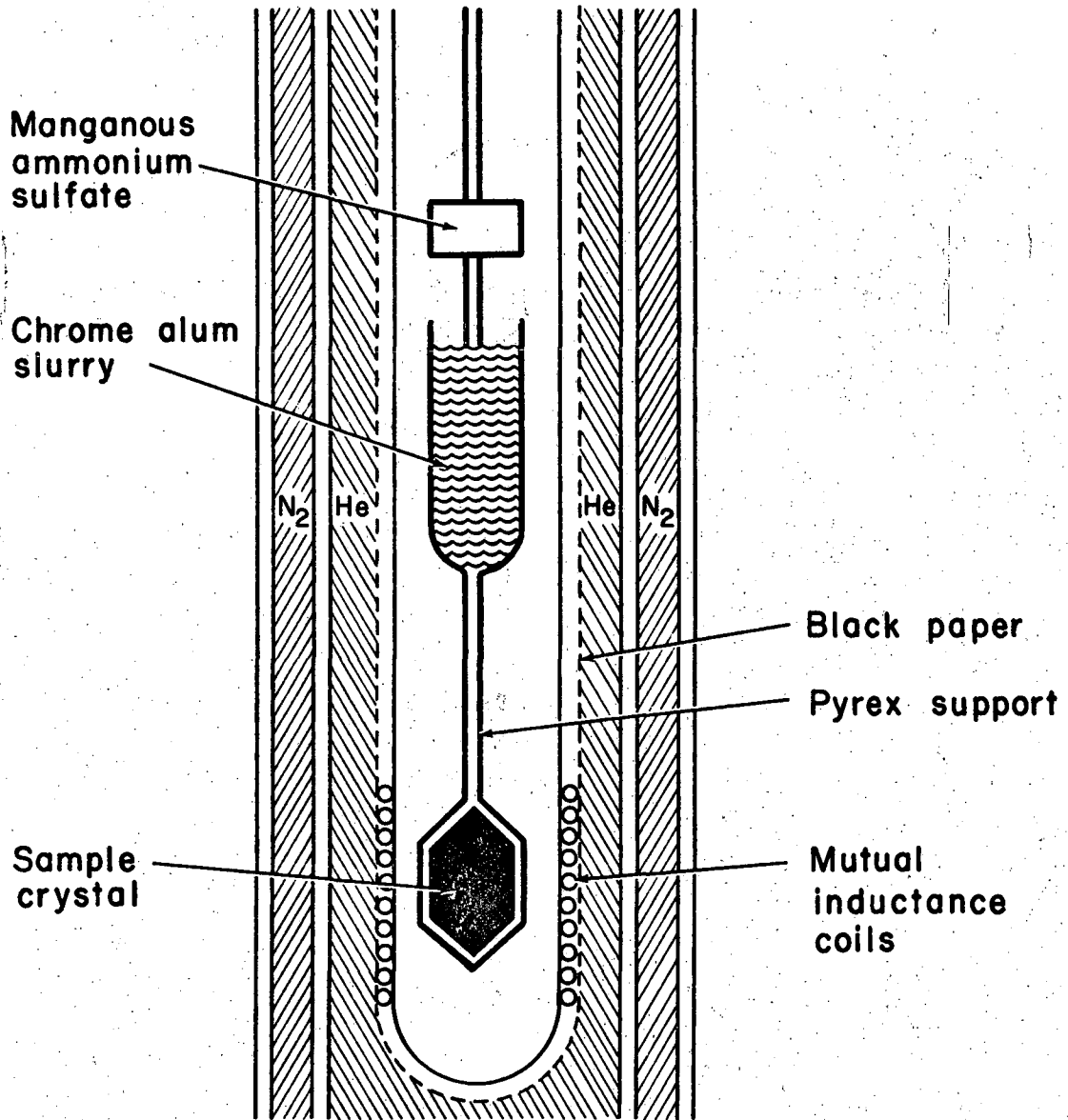
Fig. 1

XBL682-1892

Fig. 2

Fig. 3

XBL682-1893

I/T (deg K)⁻¹
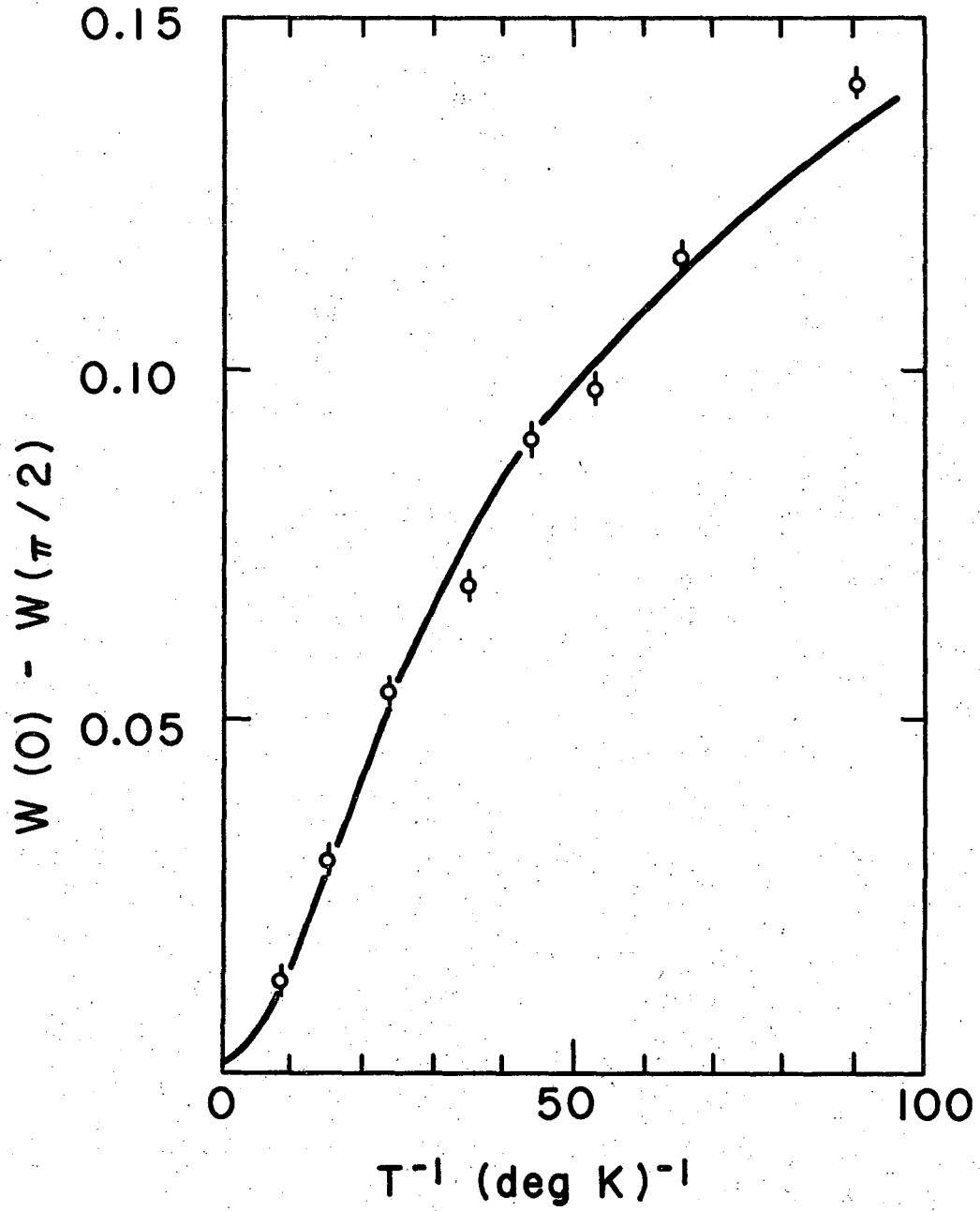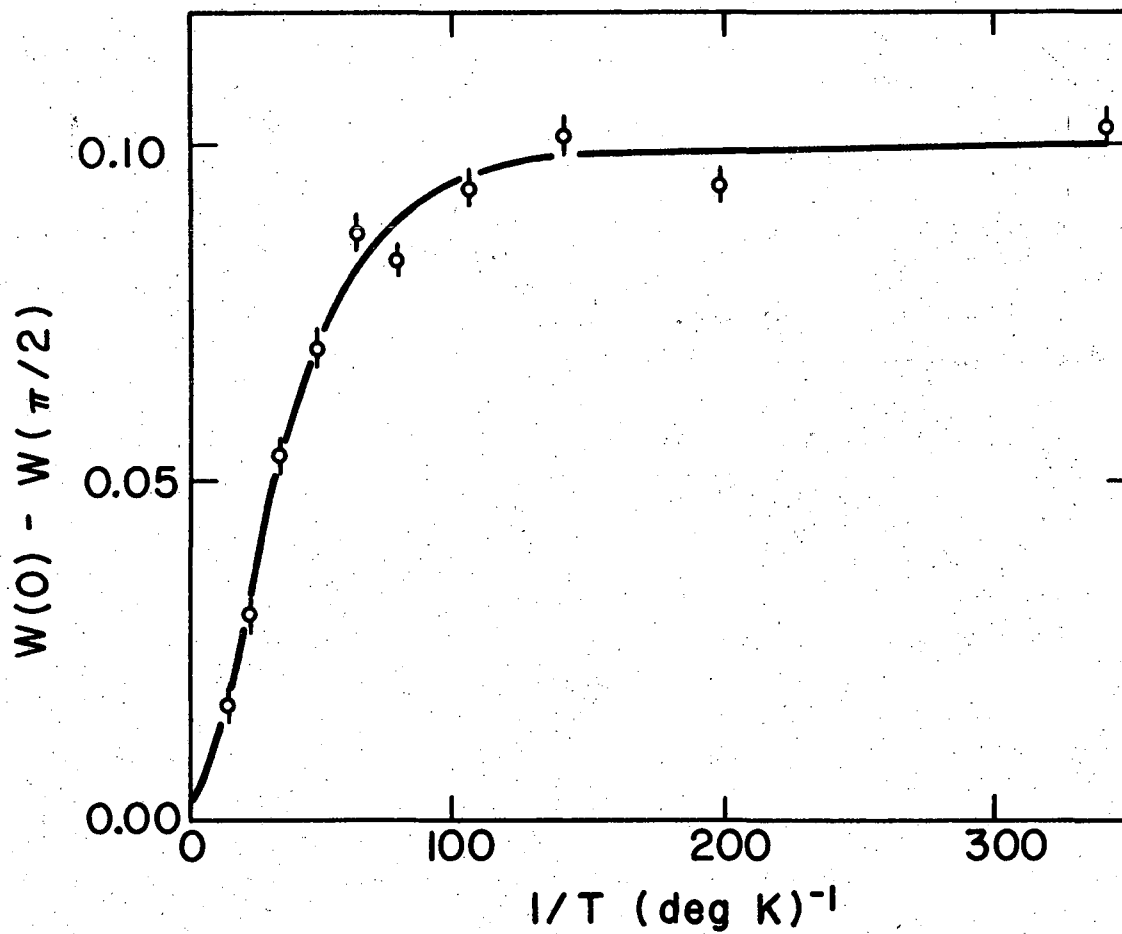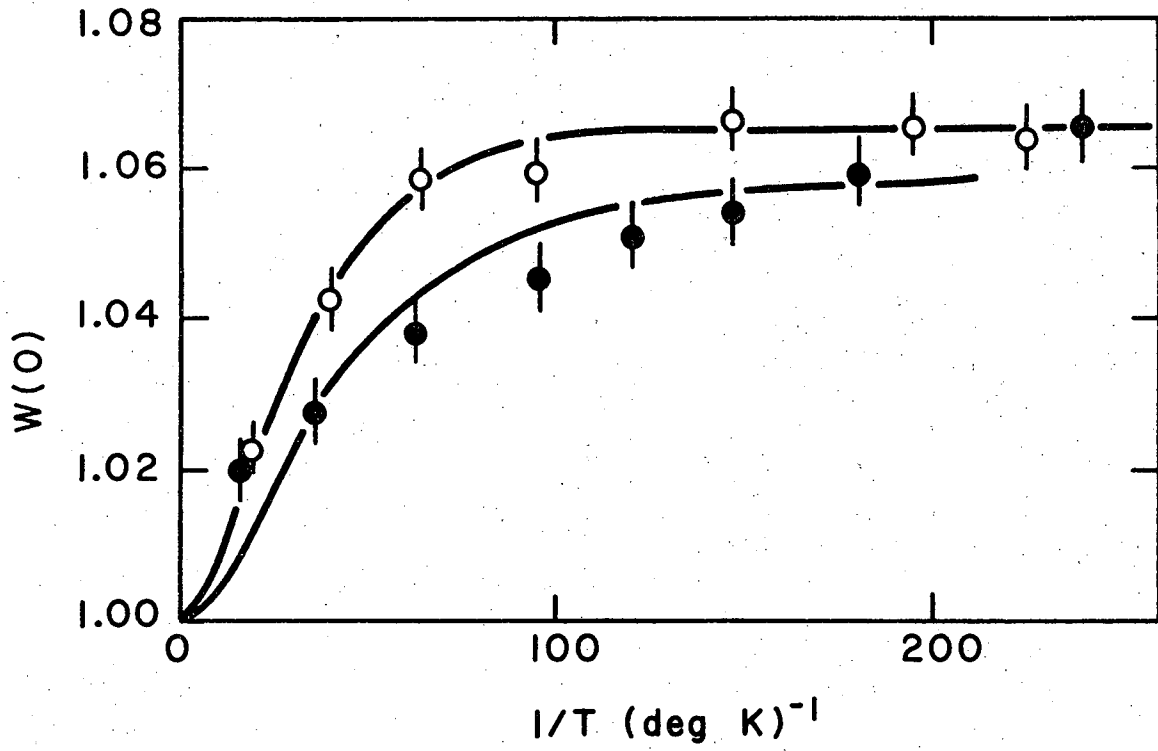
XBL682-1894

Fig. 4

XBL682-1895

Fig. 5